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(54)【発明の名称】 熱可塑性樹脂組成物及びそれからなるシール材

(57)【要約】

【課題】 ポリエステルやポリプロピレン等との易開封性に優れ、低湿度でも帯電防止効果を有し、且つ帯電防止性効果の持続性に優れたシール材を開発することにある。

【解決手段】 エチレン・ビニルエステル共重合体

(A)、低結晶性もしくは非晶性のエチレン・ α -オレフィン共重合体(B)及び分子内に水酸基を3個以上持つ分子量400以下の化合物を含むエチレン・不飽和カルボン酸共重合体のカリウムイオンアイオノマー(C)とからなることを特徴とするポリエステルやポリプロピレン等との易開封性に優れ、低湿度でも帯電防止効果を有し、且つ帯電防止性効果の持続性に優れたシール材として好適な熱可塑性樹脂組成物を提供するものである。

【特許請求の範囲】

【請求項1】エチレン・ビニルエステル共重合体

(A)、低結晶性もしくは非晶性のエチレン・ α -オレフィン共重合体(B)及び分子内に水酸基を3個以上持つ分子量400以下の化合物を含むエチレン・不飽和カルボン酸共重合体のカリウムイオンアイオノマー(C)とからなることを特徴とする熱可塑性樹脂組成物。

【請求項2】熱可塑性樹脂組成物における、エチレン・ビニルエステル共重合体(A)が20～60重量%、低結晶性もしくは非晶性のエチレン・ α -オレフィン共重合体(B)が20～60重量%及び分子内に水酸基を3個以上持つ分子量400以下の化合物を含むエチレン・不飽和カルボン酸共重合体のカリウムイオンアイオノマー(C)が1～40重量%の範囲にあることを特徴とする請求項1記載の熱可塑性樹脂組成物。

【請求項3】エチレン・ビニルエステル共重合体(A)が酢酸ビニル含有量3～25重量%のエチレン・酢酸ビニル共重合体である請求項1もしくは2に記載の熱可塑性樹脂組成物。

【請求項4】分子内に水酸基を3個以上持つ分子量400以下の化合物を含むエチレン・不飽和カルボン酸共重合体のカリウムイオンアイオノマー(C)がカリウムイオンによる中和度が70モル%以上である請求項1もしくは2に記載の熱可塑性樹脂組成物。

【請求項5】熱可塑性樹脂組成物が、更に粘着付与剤(D)を含有していることを特徴とする請求項1記載の熱可塑性樹脂組成物。

【請求項6】熱可塑性樹脂組成物における粘着付与剤(D)の含有量が3～30重量%である請求項5記載の熱可塑性樹脂組成物。

【請求項7】基材(E)の少なくとも片面に、エチレン・ビニルエステル共重合体(A)、低結晶性もしくは非晶性のエチレン・ α -オレフィン共重合体(B)及び分子内に水酸基を3個以上持つ分子量400以下の化合物を含むエチレン・不飽和カルボン酸共重合体のカリウムイオンアイオノマー(C)との熱可塑性樹脂組成物からなる熱融着層を備えたことを特徴とするシール材。

【請求項8】熱融着層を構成する熱可塑性樹脂組成物が、更に粘着付与剤(D)を含有していることを特徴とする請求項7記載のシール材。

【発明の詳細な説明】

【0001】

【発明の技術分野】本発明は、半永久的でしかも低湿度下での帯電防止性に優れた熱可塑性樹脂組成物及び当該組成物からなる熱融着層を備えたシール材、更に詳しくはポリエチレンテレフタレート、ポリエチレンナフタレート等のポリエステル及びポリプロピレンとの易開封性に優れたシール材に関する。

【0002】

【発明の技術的背景】各種飲食品や医薬品の容器とし

て、易開封性の熱融着層を備えたシール材でシールされた熱可塑性樹脂製容器が広く使用されている。このようなシール材には、ヒートシール温度幅が広く且つ適度なシール強度(剥離強度)が得られるとともに、容易に開封できる機能が求められている。そして、容器の材質や大きさ、被包装物等によって、要求されるシール強度が異なるため、種々のシール材が提案され使用されている。

【0003】ところで、このようなシール材は用途によってはフィルム表面の帯電防止性が要求されることがあり、そのため帯電防止剤が配合されている。しかしながら、シール材に帯電防止剤を配合すると、帯電防止剤がシール材の表面にブリードアウトしシール強度が低下したり、フィルム成形時にダイリップに目やに等が析出し頻繁に生産ラインを停止してダイを清掃する必要がある等の問題があった。

【0004】かかる問題点を解決する方法として、カリウムイオンアイオノマーからなる組成物を用いた積層フィルムが非帯電性に優れていることが提案されている(特開平8-267671号公報)。しかしながら、かかる積層フィルムはシール材としては万能ではなく、ポリエステルやポリプロピレン等とのシール性には劣るという問題がある。

【0005】

【発明が解決しようとする課題】そこで本発明者等は、ポリエステルやポリプロピレン等との易開封性に優れ、低湿度でも帯電防止効果を有し、且つ帯電防止性効果の持続性に優れたシール材を開発すべく種々検討した。

【0006】

【課題を解決するための手段】

【発明の概要】本発明は、エチレン・ビニルエステル共重合体(A)、低結晶性もしくは非晶性のエチレン・ α -オレフィン共重合体(B)及び分子内に水酸基を3個以上持つ分子量400以下の化合物を含むエチレン・不飽和カルボン酸共重合体のカリウムイオンアイオノマー(C)(以下、「アイオノマー(C)」と略称する場合がある。)とからなることを特徴とするポリエステルやポリプロピレン等との易開封性に優れ、低湿度でも帯電防止効果を有し、且つ帯電防止性効果の持続性に優れたシール材として好適な熱可塑性樹脂組成物に関する。更に好ましくは、熱可塑性樹脂組成物における、エチレン・ビニルエステル共重合体(A)が20～60重量%、低結晶性もしくは非晶性のエチレン・ α -オレフィン共重合体(B)が20～60重量%及びアイオノマー(C)が1～40重量%の範囲にあることを特徴とする熱可塑性樹脂組成物に関する。

【0007】本発明の他の態様は、エチレン・ビニルエステル共重合体(A)、低結晶性もしくは非晶性のエチレン・ α -オレフィン共重合体(B)及びアイオノマー(C)に加え、粘着付与剤(D)を含有してなる熱可塑性

樹脂組成物に関する。

【0008】又、本発明は、基材(E)の少なくとも片面に、エチレン・ビニルエステル共重合体(A)、低結晶性もしくは非晶性のエチレン・ α -オレフィン共重合体(B)及びアイオノマー(C)との熱可塑性樹脂組成物からなる熱融着層を備えたことを特徴とするシール材に関する。

【0009】

【発明の具体的説明】エチレン・ビニルエステル共重合体(A)

本発明の熱可塑性樹脂組成物を構成するエチレン・ビニルエステル共重合体(A)は、エチレンと酢酸ビニルやプロピオン酸ビニル等のビニルエステルとの共重合体であって、共重合体中に存在するビニルエステル含量が通常3~30重量%、好ましくは5~15重量%の範囲にある。ビニルエステル含量がこの範囲内にあると、ヒートシール強度とイージーピール性とのバランスを良好に保ち、かつ食品充填時に必要な耐熱性を有しているため好都合である。また、この共重合体のメルトフローレート(MFR)は、JIS 6730に準拠して測定した値が、通常1~40、好ましくは1.5~30(g/10分)であると、良好な押出性と包装フィルムとして用いる場合に十分な機械的強度を得る上で望ましい。このエチレン・ビニルエステル共重合体(A)は熱融着層として用いる場合に低温ヒートシール性を付与する効果がある。

【0010】低結晶性もしくは非晶性のエチレン・ α -オレフィン共重合体(B)

本発明の熱可塑性樹脂組成物を構成する低結晶性もしくは非晶性のエチレン・ α -オレフィン共重合体(B)は、エチレンとプロピレン、1-ブテン、1-ペンテン、1-ヘキセン、4-メチル-1-ペンテン、1-オクテン等の炭素数3~10の α -オレフィンとのランダム共重合体で、通常X線による結晶化度が通常30%以下、好ましくは25%以下、密度が0.86~0.92g/cm³、好ましくは0.87~0.91g/cm³、MFR(ASTM D1238 荷重2160g、温度190℃)が0.2~30g/10分、好ましくは1~10g/10分、DSCによる融点が100℃以下、好ましくは80℃以下の範囲にある。

【0011】分子内に水酸基を3個以上持つ分子量400以下の化合物を含むエチレン・不飽和カルボン酸共重合体のカリウムイオンアイオノマー(C)

本発明の熱可塑性樹脂組成物を構成するアイオノマー(C)を構成するエチレン・不飽和カルボン酸共重合体のカリウムイオンアイオノマーは、エチレン・不飽和カルボン酸共重合体をカリウムイオンで中和、好ましくは60%以上、特に好ましくは70%以上であり、またカリウムイオンの総量が、カリウムイオンアイオノマー1kg当たり0.4~4モル、とくに0.6~2モルの範

囲にあることが、非帯電性及びフィルム物性を考慮すると好ましい。勿論このようなカリウムイオンアイオノマーとしては、例えばエチレン・不飽和カルボン酸エステル共重合体をケン化することによっても製造することができる。またかかるカリウムイオンアイオノマーのMFR(190℃、2160g荷重)は、通常0.01~1000g/10分、好ましくは0.1~100g/10分の範囲にある。

【0012】前記カリウムイオンアイオノマーのベースポリマーとなるエチレン・不飽和カルボン酸共重合体は、エチレン及び不飽和カルボン酸、及び任意に他の単量体成分を共重合したものであって、通常、エチレンが60~90重量%、好ましくは70~88重量%、不飽和カルボン酸が10~40重量%、好ましくは12~30重量%、他の単量体成分が0~30重量%、好ましくは0~20重量%の割合でランダム共重合されているものである。またこのようなランダム共重合体とともに、エチレン含有量が上記のものより多く、したがって不飽和カルボン酸含有量が上記のものより少ない同様のランダム共重合体を併用してもよい。

【0013】かかる不飽和カルボン酸としては、アクリル酸、メタクリル酸、フマル酸、マレイン酸モノメチル、マレイン酸モノエチル、無水マレイン酸などを例示することができるが、特にアクリル酸又はメタクリル酸が望ましい。また任意共重合成分である他の単量体成分としては、不飽和カルボン酸のエステル、例えばアクリル酸メチル、アクリル酸エチル、アクリル酸イソプロピル、アクリル酸イソブチル、アクリル酸nブチル、アクリル酸2-エチルヘキシル、メタクリル酸メチル、メタクリル酸イソブチルなど、あるいはビニルエステル、例えば酢酸ビニル、プロピオン酸ビニルなどを例示することができる。このような共重合体は、高圧法ポリエチレンと同様の方法で、高温高圧下で各重合成分をラジカル共重合することによって得ることができる。

【0014】エチレン・不飽和カルボン酸共重合体のカリウムイオンアイオノマーに含まれる分子内に水酸基を3個以上持つ分子量400以下の化合物は、炭素、水素及び酸素のみから構成される化合物であってもよく、また炭素、水素、酸素の他にさらに窒素のようなヘテロ原子を含有するものであってもよい。これらは分子量が400以下、好ましくは80~300であって、室温で液体状であっても固体状であってもよい。分子量が400を越えるものを用いても改良効果は小さい。

【0015】このような化合物の具体例としては、グリセリン、ジグリセリン、トリメチロールプロパン、1,1,1-トリス(ヒドロキシメチル)エタン、2,2-ジ(ヒドロキシメチル)-1,3-プロパンジオール、ソルビトール、1,3,5-トリヒドロキシベンゼン、1,3,5-トリヒドロキシ安息香酸、トリス(ヒドロキシメチル)アミノプロパン、N,N,N',N'-テ

トラキス(2-ヒドロキシエチル)エチレンジアミン、N, N, N', N' -テトラキス(2-ヒドロキシプロピル)エチレンジアミンなどが例示できる。これらの中では、グリセリンまたはトリメチロールプロパンの如き脂肪族多価アルコールを用いるのがもっとも好ましい。

【0016】これら分子内に水酸基を3個以上持つ分子量400以下の化合物の含まれる量は、カリウムイオンアイオノマー成分100重量部当たり通常0.1から30重量部、好ましくは1~20重量部である。分子内に水酸基を3個以上持つ分子量400以下の化合物の含まれる量が上記範囲より少ない場合は非帯電性の改良効果が充分でない場合がある。又、分子内に水酸基を3個以上持つ分子量400以下の化合物の配合量が上記範囲より多くなると、配合物の調製が困難となり、又、ブリードによる表面汚染が起こりやすくなるなど好ましくない。

【0017】粘着付与剤(D)

本発明の熱可塑性樹脂組成物を構成する粘着付与剤(D)としては、クマロン・インデン樹脂、p-第三ブチルフェノール・アセチレン樹脂、フェノール・ホルムアルデヒド樹脂、テルペン・フェノール樹脂及びキシレン・ホルムアルデヒド樹脂等のフェノール系樹脂、β-ビネン樹脂、α-ビネン樹脂、ジベンテンベース樹脂及びスチレン変成テルペン樹脂、合成ポリテルペン樹脂等のテルペン系樹脂、極性基を有しないテルペン樹脂、芳香族系炭化水素樹脂、脂肪族系炭化水素樹脂、脂肪族系環状炭化水素樹脂、脂肪族・脂環族系石油樹脂、脂肪族・芳香族系石油樹脂及び水素添加炭化水素樹脂等の石油系炭化水素樹脂、ロジンのペンタエリスリトール・エステル、ロジンのグリセロール・エステル、水素添加ロジン、水素添加ロジンのペンタエリスリトール・エステル、水素添加ロジンのメチル・エステル及び水素添加ロジンのトリエチレングリコール・エステル等のロジン誘導体等が挙げられる。

【0018】これらの中でも、極性基を有しないテルペン系樹脂及び極性基を有しない石油系炭化水素樹脂が熱安定性に優れるので好ましく、更に水素添加、好ましくは80%以上、特に95%以上水素添加した極性基を有しない石油系炭化水素樹脂が加工性の面でフィッシュアイ(FE)など劣化物等の発生が少なく外観的にきれいなフィルムを製膜できる安定面に優れるので好ましい。

【0019】熱可塑性樹脂組成物

本発明の熱可塑性樹脂組成物は、エチレン・ビニルエステル共重合体(A)、低結晶性もしくは非晶性のエチレン・α-オレフィン共重合体(B)及びアイオノマー(C)とからなり、好ましくは熱可塑性樹脂組成物における、エチレン・ビニルエステル共重合体(A)が20~60重量%、更に好ましくは25~40重量%、低結晶性もしくは非晶性のエチレン・α-オレフィン共重合体

(B)が20~60重量%、更に好ましくは25~40重量%、及びアイオノマー(C)が1~40重量%、更に好ましくは10~30重量%の範囲にある。

【0020】エチレン・ビニルエステル共重合体(A)が20重量%未満では、ヒートシール強度が十分に発揮できない虞があり、一方、60重量%を越えるとボイル適正など耐熱性の面で劣る虞がある。エチレン・α-オレフィン共重合体(B)が20重量%未満では低温ヒートシール性とヒートシール強度の面で十分な性能が発揮できない虞があり、一方、60重量%を越えるとヒートシール強度が十分に発揮できない虞がある。アイオノマー(C)が20重量%未満では十分な帯電防止効果が発揮できない虞があり、一方、60重量%を越えるとヒートシール強度が十分に発揮できない虞がある。

【0021】本発明の熱可塑性樹脂組成物は、上記3成分に加え、粘着付与剤(D)を含有、好ましくは3~30重量%、更に好ましくは更に好ましくは10~25重量%含有させると重量%含有させると、易開封性シーラントとしてのヒートシール強度の安定性が改良される。

【0022】本発明の熱可塑性樹脂組成物は、上記エチレン・ビニルエステル共重合体(A)、低結晶性もしくは非晶性のエチレン・α-オレフィン共重合体(B)、アイオノマー(C)及び必要に応じて粘着付与剤(D)を上記範囲でヘンシェルミキサー、V-ブレンダー、リボンブレンダー、タンブラーミキサー等で混合する方法、混合後更に単軸押出機、多軸押出機、バンバリーミキサー等で熔融混練する方法等により得られる。

【0023】本発明の熱可塑性樹脂組成物には、エチレン・ビニルエステル共重合体(A)、低結晶性もしくは非晶性のエチレン・α-オレフィン共重合体(B)、アイオノマー(C)及び粘着付与剤(D)に夫々別個に、あるいは組成物を製造する際に、本発明の目的を損わない範囲で、通常用いられる酸化防止剤、耐候安定剤、帯電防止剤、防曇剤等の添加剤を必要に応じて配合することができる。特に、アンチブロッキング剤、スリップ剤を配合することによって、押出加工時、ラミネート加工時、包装作業時等における加工性や作業性を向上させることができる。

【0024】アンチブロッキング剤としては、シリカ、タルク、クレー、ゼオライト等の無機質粉末を挙げることができる。スリップ剤としては、ステアリン酸アミド、オレイン酸アミド、パルミチン酸アミド、エルカ酸アミド等の脂肪酸アミド；ステアリン酸、オレイン酸、エルカ酸など脂肪酸のメチレンビスアミドやエチレンビスアミドなどの脂肪酸ビスアミド；ポリエチレングリコールやポリプロピレングリコール等のポリアルキレングリコール；水添ひまし油；脂肪酸金属塩等をあげることができる。これらアンチブロッキング剤やスリップ剤は、熱可塑性樹脂組成物中に性能面で問題ない範囲で配合することが好ましい。

【0025】基材(E)

本発明のシール材と積層される基材(E)は、通常、包装材料として使用されている種々材料、例えば、ポリエチレンフィルム、ポリプロピレンフィルム、ポリブテンフィルム及びポリメチルペンテンフィルム等のポリオレフィンフィルム、ポリエチレンテレフタレートフィルム及びポリカーボネートフィルム等のポリエステルフィルム、ナイロンフィルム、ポリスチレンフィルム、ポリ塩化ビニルフィルム、ポリ塩化ビニリデンフィルム、ポリビニルアルコールフィルム、エチレン・ビニルアルコール共重合体フィルム、ポリメチルメタクリレートフィルム、エチレン・酢酸ビニル共重合体フィルム等の熱可塑性樹脂製フィルム、アルミニウム箔、紙等が挙げられる。かかる熱可塑性樹脂製フィルムからなる基材は無延伸であっても一軸あるいは二軸延伸フィルムであっても良い。勿論、基材は1層でも2層以上としても良い。

【0026】シール材

本発明のシール材は、基材(E)の少なくとも片面に上記熱可塑性樹脂組成物からなる熱融着層を積層した積層体である。かかるシール材を製造する方法としては、種々公知の方法、例えば熱可塑性樹脂組成物を用いてTダイフィルム成形機、インフレーションフィルム成形機等を用いて予めフィルムにした後、基材(E)と接着剤を介してあるいは介さずに貼り合せる方法、基材(E)に押出機を用いて熱可塑性樹脂組成物を押しラミネートする方法、基材となる熱可塑性樹脂と熱融着層となる熱可塑性樹脂組成物とを共押し成形してシール材とする方法等が例示できる。

【0027】シール材における熱可塑性樹脂組成物からなる熱融着層の厚さは、通常3~1000 μ m、好ましくは5~100 μ mの範囲にある。又、熱融着層の補強層として帯電防止剤であるアイオノマー(C)を含まない熱可塑性樹脂組成物を用いることにより、熱融着層の厚さを2 μ m程度以上と薄くすることができる。その場合補強層の厚さは通常18~998 μ mの範囲にある。補強層は1層あるいは2層以上とすることもできる。

【0028】

【発明の効果】本発明の熱可塑性樹脂組成物は、熱融着層として用いた場合にポリエステルやポリプロピレン等との易開封性に優れ、低湿度でも帯電防止効果を有し、且つ帯電防止性効果の持続性に優れており、シール材の熱融着層として好適に用い得る。又、熱融着層を得る際にも、従来主に、帯電防止剤が引き起していた溶融押し・製膜加工時の冷却ロールの汚れを少なくすることができ、また、この熱融着層を基材である他の材料とラミネート加工する場合にも、加工機のロール汚れを少なくすることができるので、成形加工作業性を向上させることができる。更に、得られる熱可塑性樹脂組成物からなるフィルムは帯電防止剤等のフィルム表面への浮き出し(ブリードアウト)が少ないので、基材に押し出しあるい

はドライラミネーションしても、ラミネート強度の低下がなく、加工時の帯電防止剤の熱による散逸がなく、より帯電性防止性能を低下させずに加工することが可能である。

【0029】又、本発明のシール材は、産業材分野の用途としてトナーなどの容器の蓋材、電子部品等の蓋材、食品、日用品等を含めあらゆる分野の包材として、フィルム表面の帯電防止性が要求される用途に、非帯電性とヒートシール機能性をバランス良くもったものとして利用できる。

【0030】

【実施例】次に実施例を挙げて本発明を更に具体的に説明するが、本発明はその要旨を越えない限りこれらの実施例に制約されるものではない。

【0031】実施例及び比較例における特性値は以下の方法で測定した。

(1) 表面固有抵抗率

(経時変化) 得られた熱融着フィルムから10×10cmの試験片を切出し、40℃のオープン中で、1日、30日及び90日間エージング処理後、オープンから取出し、20℃、相対湿度65%の雰囲気中で24時間放置した後、アドバンテスト社製デジタル超高抵抗/微量電流計(R8340A)とレジスチビティ・チェンバ(R12704)を用いて、表面固有抵抗値を測定した。

(湿度依存性) 得られた熱融着フィルムから10×10cmの試験片を切出し、40℃相対湿度60%、45%、35%の雰囲気中のオープン中で、1日間エージング処理後、オープンから取出し、30℃でエージング処理の相対湿度と同じ60%、45%、35%の雰囲気中で、上記方法で表面固有抵抗値を測定した。

(2) 非帯電性

得られた熱融着フィルムからA4サイズの試験片を切出し、40℃のオープン中で、1日、30日及び90日間エージング処理後、オープンから取出し、23℃、相対湿度60%で24時間放置した。処理した試験片を両手で持って頭で10回こすった後、試験片をテーブルに置いたたばこの灰に近づけ、灰が試験片に引き寄せられた距離を測定し、評価した。0cmまで近づけても灰の付着しない試験片を◎、0~1cmまで近づけてほんのわずかに灰が付着した試験片を○、6cmまで近づけた時点で灰が付着した試験片を×とした。

(3) ヒートシール性: 得られた熱融着フィルムと厚さ12 μ m二軸延伸ポリエチレンテレフタレートフィルム(PET)のアンカーコートした面を厚さ20 μ mの高圧法低密度ポリエチレンを用いて押しラミネーション法により積層フィルムを用意した。次いで得られた積層フィルムの熱融着フィルム面と厚さ300 μ mのポリプロピレンシート(東セロPPシートT-T)面とを重ね合せ、シールバー5mmのヒートシーラーを使用して、所定の温度で、0.2MPaの圧力で1秒間ヒートシールし

た放冷した。これから、15mm幅の試験片を切り取りクロスヘッド速度500mm/分でヒートシール部を剥離し、その強度をヒートシール強度とした。

【0032】又、本実施例で使用した原料は次の通りである。

(1) エチレン・酢酸ビニル共重合体(EVA)：酢酸ビニル含量10重量%、MFR9g/10分、融点94℃

(2) エチレン・1-ブテン共重合体(EBR)：1-ブテン含量10重量%、密度0.88g/cm³、融点68℃、MFR4.0g/10分

(3) 脂肪族系炭化水素樹脂：荒川化学(株)製品(商品名アルコン)、環球法軟化点115℃

(4) グリセリンを10wt%含有するエチレン・アクリル酸共重合体カリウムアイオノマー樹脂組成物(アイオノマー組成物)

ベースポリマー：エチレン・アクリル酸共重合体(アクリル酸含量21重量%)

金属カチオン源：カリウム

中和度：80モル%

金属カチオン含有量：2.3モル/kgアイオノマー

MFR：0.6g/10分

グリセリン(分子量92)

【0033】実施例1

EVA：36重量%とEBR：36重量%、脂肪族系炭化水素樹脂：18重量%及びアイオノマー組成物：10重量%をドライブレンドした熱可塑性組成物(熱融着層)を押出機に供給し、Tダイ法によって熱融着フィルムを得た。フィルムの厚みは30μmであった。得られた熱融着フィルムの評価結果を表1に示す。

【0034】実施例2

EVA：34重量%とEBR：34重量%、脂肪族系炭化水素樹脂：17重量%及びアイオノマー組成物：15重量%をドライブレンドした熱可塑性組成物(熱融着層)を押出機に供給し、Tダイ法によって熱融着フィルム*

*ムを得た。フィルムの厚みは30μmであった。得られた熱融着フィルムの評価結果を表1に示す。

【0035】実施例3

EVA：32重量%とEBR：32重量%、脂肪族系炭化水素樹脂：16重量%及びアイオノマー組成物：20重量%をドライブレンドした熱可塑性組成物(熱融着層)を押出機に供給し、Tダイ法によって熱融着フィルムを得た。フィルムの厚みは30μmであった。得られた熱融着フィルムの評価結果を表1に示す。

【0036】実施例4

EVA：40重量%とEBR：40重量%及びアイオノマー組成物：20重量%をドライブレンドした熱可塑性組成物(熱融着層)を押出機に供給し、Tダイ法によって熱融着フィルムを得た。フィルムの厚みは30μmであった。得られた熱融着フィルムの評価結果を表1に示す。

【0037】比較例1

EVA：40重量%とEBR：40重量%及び脂肪族系炭化水素樹脂：20重量%をドライブレンドした熱可塑性組成物(熱融着層)を押出機に供給し、Tダイ法によって熱融着フィルムを得た。フィルムの厚みは30μmであった。得られた熱融着フィルムの評価結果を表1に示す。

【0038】

【表1】

評価項目	実施例1	実施例2	実施例3	実施例4	比較例1
ヒートシール強度 (N/10mm)	140℃	5.0	7.5	5.0	7.5
	150℃	5.5	8.5	5.0	8.0
	160℃	12.5	15.5	12.0	12.5
	試験方法	JIS K 6804	JIS K 6804	JIS K 6804	JIS K 6804
熱融着層の厚さ (μm)	40℃×10分	1.5×10 ⁻¹²	2.0×10 ⁻¹²	2.5×10 ⁻¹¹	1.5×10 ⁻¹¹
	40℃×30分	2.0×10 ⁻¹²	1.0×10 ⁻¹²	0.1×10 ⁻¹¹	0.5×10 ⁻¹¹
	40℃×60分	5.5×10 ⁻¹²	2.0×10 ⁻¹²	0.5×10 ⁻¹¹	1.5×10 ⁻¹¹
熱融着層の厚さ (μm)	200℃	—	—	2.1×10 ⁻¹²	5.0×10 ⁻¹²
	450℃	—	—	1.5×10 ⁻¹²	0.7×10 ⁻¹²
	550℃	—	—	0.5×10 ⁻¹¹	1.0×10 ⁻¹¹
熱融着層の厚さ (μm)	40℃×10分	0-5	0	0	H
	40℃×30分	0-5	0	0	H
	40℃×60分	0-5	0	0	H

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(54) THERMOPLASTIC RESIN COMPOSITION AND SEALING MATERIAL USING THE SAME

(57)Abstract:

PROBLEM TO BE SOLVED: To develop a sealing material which has an excellent easy-to-open performance with polyester or polypropylene, an antistatic effect in a low moisture atmosphere and a superior continuance of the effect.

SOLUTION: The thermoplastic resin composition consists of (A) an ethylene-vinyl ester copolymer, (B) a low-crystalline or amorphous ethylene- α -olefin copolymer and (C) a potassium ion ionomer of an ethylene-unsaturated carboxylic acid copolymer containing a compound having three or more hydroxy groups in a molecule and a molecular weight of 400 or less. This composition is suited for a sealing material which has an excellent easy-to-open performance with polyester or polypropylene, an antistatic effect in a low moisture atmosphere and a superior continuance of the effect.

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CLAIMS

[Claim(s)]

[Claim 1] A thermoplastics constituent characterized by consisting of a potassium ion ionomer (C) of an ethylene and an unsaturated-carboxylic-acid copolymer containing a with a molecular weight of 400 or less which has three or more hydroxyl groups in ethylene, alpha-olefine copolymer (B), and intramolecular of an ethylene vinyl ester copolymer (A), low crystallinity, or amorphism nature compound.

[Claim 2] A thermoplastics constituent according to claim 1 characterized by a potassium ion ionomer (C) of an ethylene and an unsaturated-carboxylic-acid copolymer with which an ethylene vinyl ester copolymer (A) contains a with a molecular weight of 400 or less compound with which ethylene and alpha-olefine copolymer of low crystallinity or amorphism nature (B) have three or more hydroxyl groups in 20 - 60 % of the weight and intramolecular 20 to 60% of the weight in a thermoplastics constituent being in 1 - 40% of the weight of a range.

[Claim 3] Claim 1 whose ethylene vinyl ester copolymer (A) is an ethylene-vinylacetate copolymer of 3 - 25 % of the weight of vinyl acetate contents, or a thermoplastics constituent given in 2.

[Claim 4] Claim 1 whenever [according / a potassium ion ionomer (C) of an ethylene and an unsaturated-carboxylic-acid copolymer containing a with a molecular weight of 400 or less which has three or more hydroxyl groups in intramolecular compound / to potassium ion neutralization / whose] is more than 70 mol %, or a thermoplastics constituent given in 2.

[Claim 5] A thermoplastics constituent according to claim 1 with which a thermoplastics constituent is characterized by containing a tackifier (D) further.

[Claim 6] A thermoplastics constituent according to claim 5 whose content of a tackifier (D) in a thermoplastics constituent is 3 - 30 % of the weight.

[Claim 7] A sealant characterized by having a heat welding layer which consists of a thermoplastics constituent with a potassium ion ionomer (C) of an ethylene and an unsaturated-carboxylic-acid copolymer containing a with a molecular weight of 400 or less which has three or more hydroxyl groups at ethylene, alpha-olefine copolymer (B), and intramolecular of an ethylene vinyl ester copolymer (A), low crystallinity, or amorphism nature at least at one side of a base material (E) compound.

[Claim 8] A sealant according to claim 7 to which a thermoplastics constituent which constitutes a heat welding layer is characterized by containing a tackifier (D) further.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention is semipermanent and relates to the sealant equipped with the heat welding layer which consists of a thermoplastics constituent which was moreover excellent in the antistatic nature under low humidity, and the constituent concerned, and the sealant which was excellent in open-easiness with polyester, such as polyethylene terephthalate and polyethylenenaphthalate, and polypropylene in more detail.

[0002]

[Background of the Invention] The container made of thermoplastics by which the seal was carried out by the sealant equipped with the heat welding layer of open-easiness as a container of various eating-and-drinking articles or drugs is used widely. While seal reinforcement (peel strength) with it is obtained by such sealant, it is asked for the function which can be opened easily. [wide and heat-sealing temperature width of face and] [moderate] And with the quality of the material of a container, magnitude, packaging goods-ed, etc., since the seal reinforcement demanded differs, various sealants are proposed and used.

[0003] By the way, depending on a use, the antistatic nature on the surface of a film may be required, therefore, as for such a sealant, the antistatic agent is blended. However, when the antistatic agent was blended with the sealant, the antistatic agent carried out bleed out on the surface of the sealant, seal reinforcement fell and there were problems, like there is the necessity of a discharge from the eyes etc. depositing in a die lip, stopping a production line frequently, and cleaning a die at the time of film shaping.

[0004] It is proposed that the laminated film using the constituent which consists of a potassium ion ionomer is excellent in un-charging nature as a method of solving this trouble (JP,8-267671,A). However, this laminated film has as a sealant the problem of being inferior in seal nature with not omnipotent but polyester, polypropylene, etc.

[0005]

[Problem(s) to be Solved by the Invention] Then, this invention person etc. examined many things that the sealant which is excellent in open-easiness with polyester, polypropylene, etc., and has the antistatic effect also with low humidity, and was excellent in the durability of the antistatic nature effect should be developed.

[0006]

[Means for Solving the Problem]

[Summary of the Invention] This invention An ethylene vinyl ester copolymer (A), The potassium ion ionomer of the ethylene and the unsaturated-carboxylic-acid copolymer containing a with a molecular weight of 400 or less which has three or more hydroxyl groups in the ethylene, alpha-olefine copolymer (B), and intramolecular of low crystallinity or amorphism nature compound (C) (It may be hereafter called "an ionomer (C)" for short.) from — it is related with a thermoplastics constituent suitable as a sealant which is excellent in open-easiness with polyester, polypropylene, etc. which are characterized by becoming, and has the antistatic effect also with low humidity, and was excellent in the durability of the antistatic nature effect. furthermore — desirable — The ethylene vinyl ester copolymer (A) in a thermoplastics constituent is related with the thermoplastics constituent with which the ethylene and alpha-olefine copolymer of low crystallinity or amorphism nature (B) are characterized by 20 – 60 % of the weight and an ionomer (C) being in 1 – 40% of the weight of a range 20 to 60% of the weight.

[0007] In addition to the ethylene, alpha-olefine copolymer (B), and the ionomer (C) of an ethylene vinyl

ester copolymer (A), low crystallinity, or amorphism nature, other modes of this invention are related with the thermoplastics constituent which comes to contain a tackifier (D).

[0008] Moreover, this invention relates to the sealant characterized by having the heat welding layer which becomes at least one side of a base material (E) from the ethylene and alpha-olefine copolymer (B) of an ethylene vinyl ester copolymer (A), low crystallinity, or amorphism nature, and a thermoplastics constituent with an ionomer (C).

[0009]

[Detailed Description of the Invention] The ethylene vinyl ester copolymer (A) which constitutes the thermoplastics constituent of ethylene vinyl ester copolymer (A) this invention is a copolymer of ethylene and vinyl ester, such as vinyl acetate and propionic-acid vinyl, and 3 - 30 % of the weight of vinyl ester contents which exist in a copolymer is usually in 5 - 15% of the weight of a range preferably. If a vinyl ester content is within the limits of this, since it keeps good the balance of heat-sealing reinforcement and easy PIRU nature and has thermal resistance required at the time of food restoration, it is convenient. Moreover, the melt flow rate (MFR) of this copolymer is JIS. The value measured based on 6730 is usually desirable, when obtaining 1-40, and mechanical strength sufficient when using that it is 1.5-30 (g / 10 minutes) preferably as good extrusion nature and a package film. This ethylene vinyl ester copolymer (A) has the effect which gives low-temperature heat-sealing nature, when using as a heat welding layer.

[0010] The ethylene and alpha-olefine copolymer (B) of the low crystallinity which constitutes the thermoplastics constituent of the ethylene and alpha-olefine copolymer (B) this invention of low crystallinity or amorphism nature, or amorphism nature By the random copolymer with alpha-olefin of the carbon numbers 3-10, such as ethylene, a propylene, 1-butene, 1-pentene, 1-hexene, 4-MECHIRU 1-pentene, and 1-octene The degree of crystallinity by the X-ray preferably 30% or less usually Usually, 25% or less, density — 0.86 - 0.92 g/cm³ — desirable — 0.87 - 0.91 g/cm³ and MFR (2160g of ASTM D1238 loads) 0.2-30g / melting point by DSC has preferably 100 degrees C or less of temperature of 190 degrees C in the range of 80 degrees C or less for 1-10g / 10 minutes for 10 minutes.
 [0011] To

intramolecular, a hydroxyl group The potassium ion ionomer of the ethylene and the unsaturated-carboxylic-acid copolymer which constitutes the ionomer (C) which constitutes the thermoplastics constituent of potassium ion ionomer (C) this invention of the ethylene and the unsaturated-carboxylic-acid copolymer containing a with a molecular weight of 400 or less which it has three or more pieces compound ethylene and an unsaturated-carboxylic-acid copolymer — potassium ion — neutralization — 60% or more preferably When that it is especially 70% or more preferably, and 0.4-4 mols per potassium ion ionomer 1kg of total amounts of potassium ion are in the range of 0.6-2 mols especially takes into consideration un-charging nature and film physical properties, it is desirable. As such [, of course] a potassium ion ionomer, it can manufacture also by saponifying ethylene and an unsaturated-carboxylic-acid ester copolymer, for example. moreover, MFR (190 degrees C, 2160g load) of this potassium ion ionomer — usually — 0.01-1000g/ — 10 minutes are in the range for 0.1-100g / 10 minutes preferably.

[0012] The monomer component of others [copolymer / the ethylene used as the base polymer of said potassium ion ionomer and / unsaturated-carboxylic-acid / arbitration / ethylene, unsaturated carboxylic acid, and] is copolymerized, ten to 40% of the weight, ethylene is carried out for 12 - 30 % of the weight, and other monomers component at 70 - 88 % of the weight, and random copolymerization of the unsaturated carboxylic acid is usually preferably carried out at 0 - 20% of the weight of a rate zero to 30% of the weight 60 to 90% of the weight. Moreover, with such a random copolymer, an ethylene content may use together more [therefore] same random copolymers with few unsaturated-carboxylic-acid contents than the above-mentioned thing than the above-mentioned thing.

[0013] As this unsaturated carboxylic acid, although an acrylic acid, a methacrylic acid, a fumaric acid, maleic-acid monomethyl, maleic-acid monoethyl, a maleic anhydride, etc. can be illustrated, especially an acrylic acid or a methacrylic acid is desirable. Moreover, as other monomer components which are arbitration copolymerization components, the ester of unsaturated carboxylic acid, for example, a methyl acrylate, an ethyl acrylate, acrylic-acid isopropyl, isobutyl acrylate, acrylic-acid n butyl, 2-ethylhexyl acrylate, a methyl methacrylate, methacrylic-acid isobutyl, etc. can illustrate vinyl ester, for example, vinyl acetate, or propionic-acid vinyl. Such a copolymer is the same method as high pressure produced polyethylene, and can be obtained by carrying out radical copolymerization of each polymerization component by the elevated-temperature high draft.

[0014] A with a molecular weight of 400 or less which has three or more hydroxyl groups in the

intramolecular contained in the potassium ion ionomer of ethylene and an unsaturated-carboxylic-acid copolymer compound may be a compound which consists of only carbon, hydrogen, and oxygen, and may contain a hetero atom still like nitrogen other than carbon, hydrogen, and oxygen. Molecular weight may be 80-300 preferably 400 or less, and these may have it at a room temperature, or may be solid-states-like. [liquid] The amelioration effect is small even if it uses that to which molecular weight exceeds 400.

[0015] As an example of such a compound, a glycerol, diglycerol, trimethylol propane, 1, 1, and 1-tris (hydroxymethyl) ethane, 2, and 2-JI (hydroxymethyl)-1,3-propanediol, A sorbitol, 1 and 3, 5-trihydroxy benzene, 1 and 3, 5-trihydroxy benzoic acid, A tris (hydroxymethyl) amino propane, N and N, N', N'-tetrakis (2-hydroxyethyl) ethylenediamine, N and N, N', N'-tetrakis (2-hydroxypropyl) ethylenediamine, etc. can be illustrated. In these, it is most desirable to use a glycerol or the aliphatic series polyhydric alcohol like trimethylol propane.

[0016] the amount in which a with a molecular weight of 400 or less which has three or more hydroxyl groups in these intramoleculars compound is contained — per potassium ion ionomer component 100 weight section — usual 0.1 to 30 weight section — it is 1 - 20 weight section preferably. When there are few amounts in which a with a molecular weight of 400 or less which has three or more hydroxyl groups in intramolecular compound is contained than the above-mentioned range, the amelioration effect of un-charging nature may not be enough. Moreover, it is not [become / the surface contamination by bleeding / preparation of a compound becomes difficult and / easy to happen] desirable if the loadings of a with a molecular weight of 400 or less which has three or more hydroxyl groups in intramolecular compound increase more than the above-mentioned range.

[0017] As a tackifier (D) which constitutes the thermoplastics constituent of tackifier (D) this invention the [coumarone-indene resin and / p-] — 3-butylphenol acetylene resin — Phenol system resin, such as phenol-formaldehyde resin, terpene phenol resin, and xylene-formaldehyde resin, beta-pinene resin, alpha-pinene resin, dipentene base resin, and styrene conversion terpene resin, Terpene system resin, such as synthetic polyterpene resin, the terpene resin which does not have a polar group, Aromatic series system hydrocarbon resin, aliphatic series system hydrocarbon resin, aliphatic series system cyclic-hydrocarbon resin, Petroleum system hydrocarbon resin, such as aliphatic series and alicycle group system petroleum resin, aliphatic series and aromatic series system petroleum resin, and hydrogenation hydrocarbon resin, The pentaerythritol ester of rosin, the glycerol ester of rosin, Rosin derivatives, such as pentaerythritol ester of hydrogenation rosin and hydrogenation rosin, methyl ester of hydrogenation rosin, and triethylene glycol ester of hydrogenation rosin, etc. are mentioned.

[0018] since the petroleum system hydrocarbon resin which does not have in these the terpene system resin and the polar group which do not have a polar group is excellent in thermal stability, it is desirable, and since hydrogenation and the petroleum system hydrocarbon resin which does not have the polar group hydrogenated 95% or more especially preferably 80% or more are further alike in respect of the stability which can produce a film with it in respect of processability and it excels, it is desirable. [there is little generating of deterioration objects, such as a fish eye (FE), etc., and beautiful in appearance]

[0019] The thermoplastics constituent of thermoplastics constituent this invention It consists of the ethylene, alpha-olefine copolymer (B), and the ionomer (C) of an ethylene vinyl ester copolymer (A), low crystallinity, or amorphism nature. The ethylene vinyl ester copolymer (A) in a thermoplastics constituent preferably 20 - 60 % of the weight, Furthermore, the ethylene and alpha-olefine copolymer of low crystallinity or amorphism nature (B) are to 25 - 40% of the weight, and 1 - 40 % of the weight (C) of ionomers is in 10 - 30% of the weight of a range still more preferably still more preferably 20 to 60% of the weight 25 to 40% of the weight preferably.

[0020] if there is a possibility that heat-sealing reinforcement cannot fully demonstrate [an ethylene vinyl ester copolymer (A)] at less than 20 % of the weight and 60 % of the weight is exceeded on the other hand — voile — there is a possibility that it may be inferior in respect of proper *****. There is a possibility that ethylene and alpha-olefine copolymer (B) cannot demonstrate engine performance sufficient in the field of low-temperature heat-sealing nature and heat-sealing reinforcement at less than 20 % of the weight, and on the other hand, when 60 % of the weight is exceeded, there is a possibility that heat-sealing reinforcement cannot fully demonstrate. There is a possibility that an ionomer (C) cannot demonstrate antistatic effect sufficient at less than 20 % of the weight, and on the other hand, when 60 % of the weight is exceeded, there is a possibility that heat-sealing reinforcement cannot fully demonstrate.

[0021] the thermoplastics constituent of this invention — the three above-mentioned component —

adding — a tackifier (D) — content — desirable — 3— if it is made to contain ten to 25% of the weight still more desirable still more preferably and weight % content of will be done 30% of the weight, the stability of the heat-sealing reinforcement as an open-easiness sealant will be improved.

[0022] The thermoplastics constituent of this invention is obtained the ethylene and alpha-olefine copolymer (B) of the above-mentioned ethylene vinyl ester copolymer (A), low crystallinity, or amorphism nature, an ionomer (C), and if needed by the method of mixing a tackifier (D) by the Henschel mixer, V-blender, the ribbon blender, a tumbler mixer, etc. in the above-mentioned range, the method of carrying out melting kneading with a single screw extruder, a multi-screw extruder, a Banbury mixer, etc. further after mixing, etc.

[0023] In case a constituent is separately manufactured, respectively to the ethylene and alpha-olefine copolymer (B) of an ethylene vinyl ester copolymer (A), low crystallinity, or amorphism nature, an ionomer (C), and a tackifier (D), additives, such as an antioxidant usually used, a weathering stabilizer, an antistatic agent, and an antifogger, can be blended with the thermoplastics constituent of this invention in the range which does not spoil the purpose of this invention if needed. The processability and workability in the time of a package etc. can be raised by blending an anti blocking agent and a slipping agent especially at the time of lamination at the time of extrusion.

[0024] As an anti blocking agent, minerals powder, such as a silica, talc, clay, and a zeolite, can be mentioned. as a slipping agent — fatty acids, such as a methylenebis amide of fatty acids, such as fatty-acid amide; stearin acid, such as octadecanamide, oleic amide, a palmitic-acid amide, and an erucic-acid amide, oleic acid, and an erucic acid, and an ethylene bis-amide, — bis— polyalkylene glycol; hydrogenation castor oil; fatty-acid metal salts, such as an amide; polyethylene glycol and a polypropylene glycol, etc. can be raised. As for these anti blocking agent or a slipping agent, it is desirable to blend in the satisfactory range in respect of the engine performance into a thermoplastics constituent.

[0025] The base material (E) by which a laminating is carried out to the sealant of base material (E) this invention usually, it is used as wrapping — various — a material, for example, a polyethylene film, — Polyolefine films, such as a polypropylene film, a polybutene film, and the poly methyl pentene film, Polyester film, such as a polyethylene terephthalate film and a polycarbonate film, A nylon film, a polystyrene film, a polyvinyl chloride film, Films made of thermoplastics, such as a polyvinylidene chloride film, a polyvinyl alcohol film, an ethylene vinyl alcohol copolymer film, a polymethylmethacrylate film, and ethylene, a vinyl acetate copolymer film, an aluminum foil, paper, etc. are mentioned. The base material which consists of this film made of thermoplastics could be extended, or may be one shaft or a biaxially oriented film. Of course, at least one layer of a base material is good also as more than two-layer.

[0026] The sealant of sealant this invention is the layered product which carried out the laminating of the heat welding layer which becomes at least one side of a base material (E) from the above-mentioned thermoplastics constituent. As a method of manufacturing this sealant, a well-known method, for example, a thermoplastics constituent, is used variously. T-die film making machine, After making it a film beforehand using a tubular blown film making machine etc., The method of sticking without minding through a base material (E) and adhesives, the method of using an extruder for a base material (E), and extruding and laminating a thermoplastics constituent, the method of carrying out co-extrusion shaping of the thermoplastics constituent used as the thermoplastics used as a base material and a heat welding layer, and making it into a sealant, etc. can be illustrated.

[0027] 3–1000 micrometers of thickness of the heat welding layer which consists of a thermoplastics constituent in a sealant are usually in the range of 5–100 micrometers preferably. Moreover, thickness of a heat welding layer can be made thin with about 2 micrometers or more by using the thermoplastics constituent which does not contain the ionomer (C) which is an antistatic agent as a reinforcement layer of a heat welding layer. In that case, the thickness of a reinforcement layer is usually in the range of 18–998 micrometers. A reinforcement layer can also be carried out to one layer or more than two-layer.

[0028]

[Effect of the Invention] The thermoplastics constituent of this invention is excellent in open-easiness with polyester, polypropylene, etc., when it uses as a heat welding layer, it has the antistatic effect also with low humidity, and is excellent in the durability of the antistatic nature effect, and can be suitably used as a heat welding layer of a sealant. Moreover, since the roll dirt of a processing machine can be conventionally lessened also when the dirt of the cooling roller at the time of the melting extrusion and film production processing which the antistatic agent had mainly caused can be lessened and it carries out the

lamination of this heat welding layer to other materials which are base materials also in case a heat welding layer is obtained, fabrication workability can be raised. furthermore, the film which consists of a thermoplastics constituent obtained — the film surfaces, such as an antistatic agent, — coming up (bleed out) — since it is few, even if it extrudes to a base material or carries out DORAIRA-Mine-SHON, it is possible to process it, without there being no fall of lamination reinforcement, and there being no dissipation by the heat of the antistatic agent at the time of processing, and reducing the electrification nature prevention engine performance more.

[0029] Moreover, the sealant of this invention can be used as an wrapping material of all fields including lid material, such as lid material of containers, such as a toner, and electronic parts, food, daily necessities, etc. as what had un-charging nature and heat-sealing functionality with sufficient balance in the use as which the antistatic nature on the surface of a film is required as a use of an industrial material field.

[0030]

[Example] Next, although an example is given and this invention is explained still more concretely, this invention is not restrained by these examples unless the summary is exceeded.

[0031] The characteristic value in an example and the example of a comparison was measured by the following methods.

(1) Cut down a 10x10cm test piece from the heat welding film obtained the rate of surface specific resistance (aging), and it is in 40-degree C oven, For one day, 30 days, and 90 days, after aging processing, after leaving it from oven for 24 hours in drawing, 20 degrees C, and the ambient atmosphere of 65% of relative humidity, the surface specific resistance value was measured using the digital ultra high resistance by ADVANTEST CORP. / minute amount ammeter (R8340A), and the REJISUCHIBITI chamber (R12704). (Humidity dependency) The 10x10cm test piece was cut down from the obtained heat welding film, and the surface specific resistance value was measured by the above-mentioned method from oven after aging processing for one day in the 60 same% as the relative humidity of drawing and 30-degree C aging processing, 45%, and 35% of ambient atmosphere in 60% of 40-degree-C relative humidity, 45%, and 35% of oven of an ambient atmosphere.

(2) The test piece of A4 size was cut down from the non-charged ***** welding film, and it was left at drawing, 23 degrees C, and 60% of relative humidity from oven after aging processing for one day, 30 days, and 90 days in 40-degree C oven for 24 hours. After having the processed test piece with both hands and rubbing 10 times with the head, it brought close to the ashes of the tobacco which put the test piece on the table, and the distance which ashes were able to draw near to a test piece was measured and evaluated. Even if close brought to 0cm, when the test piece to which the test piece to which ashes do not adhere was brought close to which to 0 and 0-1cm, and ashes adhered only slightly was close brought to 0 and 6cm, the test piece to which ashes adhered was made into x.

(3) Heat-sealing nature : the laminated film was prepared for the field as for which the obtained heat welding film and 12-micrometer biaxial-stretching polyethylene terephthalate film (PET) in thickness carried out the anchor coat with the extrusion lamination process using high pressure process low density polyethylene with a thickness of 20 micrometers. subsequently, superposition and a seal bar 5mm heat sealer were used, and the heat welding film plane of a laminated film and the polypropylene sheet (TOH CELLO PP sheet T-T) side with a thickness of 300 micrometers which were acquired were heat sealed for 1 second by the pressure of 0.2MPa with a predetermined temperature — it cooled radiationally. From now on, the test piece of 15mm width of face was cut off, the heat-sealing section was exfoliated in a part for 500mm/in crosshead speed, and the reinforcement was made into heat-sealing reinforcement.

[0032] Moreover, the raw material used by this example is as follows.

(1) ethylene-vinylacetate copolymer (EVA): — MFR4.0g / [10 % of the weight of (2) ethylene and 1-butene copolymer (EBR):1-butene contents with 10 % of the weight of vinyl acetate contents, 10 minutes / MFR9g / minutes /, and a melting point of 94 degrees C, density 0.88 g/cm³, the melting point of 68 degrees C, and] 115 degrees-C (4) glycerol of 10-minute aliphatic-series [(3)] system hydrocarbon resin:Arakawa Chemistry product (trade name Al Cong) and ring and ball method softening temperatures 10wt(s)% — the ethylene acrylic-acid copolymer potassium ionomer resin constituent (ionomer constituent) to contain

Base polymer: Ethylene acrylic-acid copolymer (21 % of the weight of acrylic-acid contents)

source of metal cation: — potassium neutralization whenever: — 80 mol % metal cation content: — 2.3-mol [/kg] ionomer MFR:0.6g /, and 10-minute glycerol (molecular weight 92)

[0033] Example 1EVA: Ionomer [36 % of the weight, EBR:36 % of the weight, aliphatic series system hydrocarbon resin:18 % of the weight, and] constituent: The thermoplastic constituent (heat welding layer) which carried out the dryblend of the 10 % of the weight was supplied to the extruder, and the heat welding film was obtained by the T-die method. The thickness of a film was 30 micrometers. The evaluation result of the obtained heat welding film is shown in a table 1.

[0034] Example 2EVA: Ionomer [34 % of the weight, EBR:34 % of the weight, aliphatic series system hydrocarbon resin:17 % of the weight, and] constituent: The thermoplastic constituent (heat welding layer) which carried out the dryblend of the 15 % of the weight was supplied to the extruder, and the heat welding film was obtained by the T-die method. The thickness of a film was 30 micrometers. The evaluation result of the obtained heat welding film is shown in a table 1.

[0035] Example 3EVA: Ionomer [32 % of the weight, EBR:32 % of the weight, aliphatic series system hydrocarbon resin:16 % of the weight, and] constituent: The thermoplastic constituent (heat welding layer) which carried out the dryblend of the 20 % of the weight was supplied to the extruder, and the heat welding film was obtained by the T-die method. The thickness of a film was 30 micrometers. The evaluation result of the obtained heat welding film is shown in a table 1.

[0036] Example 4EVA: Ionomer [40 % of the weight, EBR:40 % of the weight, and] constituent: The thermoplastic constituent (heat welding layer) which carried out the dryblend of the 20 % of the weight was supplied to the extruder, and the heat welding film was obtained by the T-die method. The thickness of a film was 30 micrometers. The evaluation result of the obtained heat welding film is shown in a table 1.

[0037] Aliphatic-series [example of comparison 1EVA:40 % of the weight, EBR:40 % of the weight, and] system hydrocarbon resin: The thermoplastic constituent (heat welding layer) which carried out the dryblend of the 20 % of the weight was supplied to the extruder, and the heat welding film was obtained by the T-die method. The thickness of a film was 30 micrometers. The evaluation result of the obtained heat welding film is shown in a table 1.

[0038]

[A table 1]

評価項目	例1	例2	例3	例4	比較例1
引張強度 MPa	140℃	5.0	1.5	5.0	1.5
	160℃	6.5	8.5	6.5	9.0
	180℃	12.5	15.5	12.0	12.5
	90℃×10分	5.0	5.0	5.0	5.0
	90℃×30分	5.0	5.0	5.0	5.0
熱安定性 %	40℃×10分	1.5×10 ⁻¹²	5.0×10 ⁻¹²	1.5×10 ⁻¹²	5.0×10 ⁻¹²
	40℃×30分	2.0×10 ⁻¹²	1.0×10 ⁻¹²	0.5×10 ⁻¹²	2.0×10 ⁻¹²
	40℃×90分	2.0×10 ⁻¹²	2.0×10 ⁻¹²	1.5×10 ⁻¹²	5.0×10 ⁻¹²
熱安定性 %	80℃×10分	—	—	2.1×10 ⁻¹²	5.0×10 ⁻¹²
	80℃×30分	—	—	1.5×10 ⁻¹²	5.0×10 ⁻¹²
	80℃×90分	—	—	0.5×10 ⁻¹²	5.0×10 ⁻¹²
熱安定性 %	40℃×10分	0-0	0	0	0
	40℃×30分	0-0	0	0	0
	40℃×90分	0-0	0	0	0

[Translation done.]

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TECHNICAL FIELD

[Field of the Invention] This invention is semipermanent and relates to the sealant equipped with the heat welding layer which consists of a thermoplastics constituent which was moreover excellent in the antistatic nature under low humidity, and the constituent concerned, and the sealant which was excellent in open-easiness with polyester, such as polyethylene terephthalate and polyethylenenaphthalate, and polypropylene in more detail.

[0002]

[Background of the Invention] The container made of thermoplastics by which the seal was carried out by the sealant equipped with the heat welding layer of open-easiness as a container of various eating-and-drinking articles or drugs is used widely. While seal reinforcement (peel strength) with it is obtained by such sealant, it is asked for the function which can be opened easily. [wide and heat-sealing temperature width of face and] [moderate] And with the quality of the material of a container, magnitude, packaging goods-ed, etc., since the seal reinforcement demanded differs, various sealants are proposed and used.

[0003] By the way, depending on a use, the antistatic nature on the surface of a film may be required, therefore, as for such a sealant, the antistatic agent is blended. However, when the antistatic agent was blended with the sealant, the antistatic agent carried out bleed out on the surface of the sealant, seal reinforcement fell and there were problems, like there is the necessity of a discharge from the eyes etc. depositing in a die lip, stopping a production line frequently, and cleaning a die at the time of film shaping.

[0004] It is proposed that the laminated film using the constituent which consists of a potassium ion ionomer is excellent in un-charging nature as a method of solving this trouble (JP,8-267671,A). However, this laminated film has as a sealant the problem of being inferior in seal nature with not omnipotent but polyester, polypropylene, etc.

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EFFECT OF THE INVENTION

[Effect of the Invention] The thermoplastics constituent of this invention is excellent in open-easiness with polyester, polypropylene, etc., when it uses as a heat welding layer, it has the antistatic effect also with low humidity, and is excellent in the durability of the antistatic nature effect, and can be suitably used as a heat welding layer of a sealant. Moreover, since the roll dirt of a processing machine can be conventionally lessened also when the dirt of the cooling roller at the time of the melting extrusion and film production processing which the antistatic agent had mainly caused can be lessened and it carries out the lamination of this heat welding layer to other materials which are base materials also in case a heat welding layer is obtained, fabrication workability can be raised. furthermore, the film which consists of a thermoplastics constituent obtained — the film surfaces, such as an antistatic agent, — coming up (bleed out) — since it is few, even if it extrudes to a base material or carries out DORAIRA-Mine-SHON, it is possible to process it, without there being no fall of lamination reinforcement, and there being no dissipation by the heat of the antistatic agent at the time of processing, and reducing the electrification nature prevention engine performance more.

[0029] Moreover, the sealant of this invention can be used as an wrapping material of all fields including lid material, such as lid material of containers, such as a toner, and electronic parts, food, daily necessities, etc. as what had un-charging nature and heat-sealing functionality with sufficient balance in the use as which the antistatic nature on the surface of a film is required as a use of an industrial material field.

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] Then, this invention person etc. examined many things that the sealant which is excellent in open-easiness with polyester, polypropylene, etc., and has the antistatic effect also with low humidity, and was excellent in the durability of the antistatic nature effect should be developed.

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MEANS

[Means for Solving the Problem]

[Summary of the Invention] This invention An ethylene vinyl ester copolymer (A), The potassium ion ionomer of the ethylene and the unsaturated-carboxylic-acid copolymer containing a with a molecular weight of 400 or less which has three or more hydroxyl groups in the ethylene, alpha-olefine copolymer (B), and intramolecular of low crystallinity or amorphism nature compound (C) (It may be hereafter called "an ionomer (C)" for short.) from — it is related with a thermoplastics constituent suitable as a sealant which is excellent in open-easiness with polyester, polypropylene, etc. which are characterized by becoming, and has the antistatic effect also with low humidity, and was excellent in the durability of the antistatic nature effect. furthermore — desirable — The ethylene vinyl ester copolymer (A) in a thermoplastics constituent is related with the thermoplastics constituent with which the ethylene and alpha-olefine copolymer of low crystallinity or amorphism nature (B) are characterized by 20 – 60 % of the weight and an ionomer (C) being in 1 – 40% of the weight of a range 20 to 60% of the weight.

[0007] In addition to the ethylene, alpha-olefine copolymer (B), and the ionomer (C) of an ethylene vinyl ester copolymer (A), low crystallinity, or amorphism nature, other modes of this invention are related with the thermoplastics constituent which comes to contain a tackifier (D).

[0008] Moreover, this invention relates to the sealant characterized by having the heat welding layer which becomes at least one side of a base material (E) from the ethylene and alpha-olefine copolymer (B) of an ethylene vinyl ester copolymer (A), low crystallinity, or amorphism nature, and a thermoplastics constituent with an ionomer (C).

[0009]

[Detailed Description of the Invention] The ethylene vinyl ester copolymer (A) which constitutes the thermoplastics constituent of ethylene vinyl ester copolymer (A) this invention is a copolymer of ethylene and vinyl ester, such as vinyl acetate and propionic-acid vinyl, and 3 – 30 % of the weight of vinyl ester contents which exist in a copolymer is usually in 5 – 15% of the weight of a range preferably. If a vinyl ester content is within the limits of this, since it keeps good the balance of heat-sealing reinforcement and easy PIRU nature and has thermal resistance required at the time of food restoration, it is convenient.

Moreover, the melt flow rate (MFR) of this copolymer is JIS. The value measured based on 6730 is usually desirable, when obtaining 1–40, and mechanical strength sufficient when using that it is 1.5–30 (g / 10 minutes) preferably as good extrusion nature and a package film. This ethylene vinyl ester copolymer (A) has the effect which gives low-temperature heat-sealing nature, when using as a heat welding layer.

[0010] The ethylene and alpha-olefine copolymer (B) of the low crystallinity which constitutes the thermoplastics constituent of the ethylene and alpha-olefine copolymer (B) this invention of low crystallinity or amorphism nature, or amorphism nature By the random copolymer with alpha-olefin of the carbon numbers 3–10, such as ethylene, a propylene, 1-butene, 1 1 pentene, 1-hexene, 4-MECHIRU 1-pentene, and 1-octene The degree of crystallinity by the X-ray preferably 30% or less usually Usually, 25% or less, density — 0.86 – 0.92 g/cm³ — desirable — 0.87 – 0.91 g/cm³ and MFR (2160g of ASTM D1238 loads) 0.2–30g / melting point by DSC has preferably 100 degrees C or less of temperature of 190 degrees C in the range of 80 degrees C or less for 1–10g / 10 minutes for 10 minutes.

[0011] To intramolecular, a hydroxyl group The potassium ion ionomer of the ethylene and the unsaturated-carboxylic-acid copolymer which constitutes the ionomer (C) which constitutes the thermoplastics constituent of potassium ion ionomer (C) this invention of the ethylene and the unsaturated-carboxylic-acid copolymer containing a with a molecular weight of 400 or less which it has

three or more pieces compound ethylene and an unsaturated-carboxylic-acid copolymer — potassium ion — neutralization — 60% or more preferably When that it is especially 70% or more preferably, and 0.4–4 mols per potassium ion ionomer 1kg of total amounts of potassium ion are in the range of 0.6–2 mols especially takes into consideration un-charging nature and film physical properties, it is desirable. As such [, of course] a potassium ion ionomer, it can manufacture also by saponifying ethylene and an unsaturated-carboxylic-acid ester copolymer, for example. moreover, MFR (190 degrees C, 2160g load) of this potassium ion ionomer — usually — 0.01–1000g/— 10 minutes are in the range for 0.1–100g / 10 minutes preferably.

[0012] The monomer component of others [copolymer / the ethylene used as the base polymer of said potassium ion ionomer and / unsaturated-carboxylic-acid / arbitration / ethylene, unsaturated carboxylic acid, and] is copolymerized, ten to 40% of the weight, ethylene is carried out for 12 – 30 % of the weight, and other monomers component at 70 – 88 % of the weight, and random copolymerization of the unsaturated carboxylic acid is usually preferably carried out at 0 – 20% of the weight of a rate zero to 30% of the weight 60 to 90% of the weight. Moreover, with such a random copolymer, an ethylene content may use together more [therefore] same random copolymers with few unsaturated-carboxylic-acid contents than the above-mentioned thing than the above-mentioned thing.

[0013] As this unsaturated carboxylic acid, although an acrylic acid, a methacrylic acid, a fumaric acid, maleic-acid monomethyl, maleic-acid monoethyl, a maleic anhydride, etc. can be illustrated, especially an acrylic acid or a methacrylic acid is desirable. Moreover, as other monomer components which are arbitration copolymerization components, the ester of unsaturated carboxylic acid, for example, a methyl acrylate, an ethyl acrylate, acrylic-acid isopropyl, isobutyl acrylate, acrylic-acid n butyl, 2-ethylhexyl acrylate, a methyl methacrylate, methacrylic-acid isobutyl, etc. can illustrate vinyl ester, for example, vinyl acetate, or propionic-acid vinyl. Such a copolymer is the same method as high pressure produced polyethylene, and can be obtained by carrying out radical copolymerization of each polymerization component by the elevated-temperature high draft.

[0014] A with a molecular weight of 400 or less which has three or more hydroxyl groups in the intramolecular contained in the potassium ion ionomer of ethylene and an unsaturated-carboxylic-acid copolymer compound may be a compound which consists of only carbon, hydrogen, and oxygen, and may contain a hetero atom still like nitrogen other than carbon, hydrogen, and oxygen. Molecular weight may be 80–300 preferably 400 or less, and these may have it at a room temperature, or may be solid-states-like. [liquid] The amelioration effect is small even if it uses that to which molecular weight exceeds 400.

[0015] As an example of such a compound, a glycerol, diglycerol, trimethylol propane, 1, 1, and 1-tris (hydroxymethyl) ethane, 2, and 2-JI (hydroxymethyl)-1,3-propanediol, A sorbitol, 1 and 3, 5-trihydroxy benzene, 1 and 3, 5-trihydroxy benzoic acid, A tris (hydroxymethyl) amino propane, N and N, N', N'-tetrakis (2-hydroxyethyl) ethylenediamine, N and N, N', N'-tetrakis (2-hydroxypropyl) ethylenediamine, etc. can be illustrated. In these, it is most desirable to use a glycerol or the aliphatic series polyhydric alcohol like trimethylol propane.

[0016] the amount in which a with a molecular weight of 400 or less which has three or more hydroxyl groups in these intramoleculars compound is contained — per potassium ion ionomer component 100 weight section — usual 0.1 to 30 weight section — it is 1 – 20 weight section preferably. When there are few amounts in which a with a molecular weight of 400 or less which has three or more hydroxyl groups in intramolecular compound is contained than the above-mentioned range, the amelioration effect of un-charging nature may not be enough. Moreover, it is not [become / the surface contamination by bleeding / preparation of a compound becomes difficult and / easy to happen] desirable if the loadings of a with a molecular weight of 400 or less which has three or more hydroxyl groups in intramolecular compound increase more than the above-mentioned range.

[0017] As a tackifier (D) which constitutes the thermoplastics constituent of tackifier (D) this invention the [coumarone-indene resin and / p-] — 3-butylphenol acetylene resin — Phenol system resin, such as phenol-formaldehyde resin, terpene phenol resin, and xylene-formaldehyde resin, beta-pinene resin, alpha-pinene resin, dipentene base resin, and styrene conversion terpene resin, Terpene system resin, such as synthetic polyterpene resin, the terpene resin which does not have a polar group, Aromatic series system hydrocarbon resin, aliphatic series system hydrocarbon resin, aliphatic series system cyclic-hydrocarbon resin, Petroleum system hydrocarbon resin, such as aliphatic series and alicycle group system petroleum resin, aliphatic series and aromatic series system petroleum resin, and hydrogenation hydrocarbon resin,

The pentaerythritol ester of rosin, the glycerol ester of rosin, Rosin derivatives, such as pentaerythritol ester of hydrogenation rosin and hydrogenation rosin, methyl ester of hydrogenation rosin, and triethylene glycol ester of hydrogenation rosin, etc. are mentioned.

[0018] since the petroleum system hydrocarbon resin which does not have in these the terpene system resin and the polar group which do not have a polar group is excellent in thermal stability, it is desirable, and since hydrogenation and the petroleum system hydrocarbon resin which does not have the polar group hydrogenated 95% or more especially preferably 80% or more are further alike in respect of the stability which can produce a film with it in respect of processability and it excels, it is desirable. [there is little generating of deterioration objects, such as a fish eye (FE), etc., and beautiful in appearance]

[0019] The thermoplastics constituent of thermoplastics constituent this invention It consists of the ethylene, alpha-olefine copolymer (B), and the ionomer (C) of an ethylene vinyl ester copolymer (A), low crystallinity, or amorphism nature. The ethylene vinyl ester copolymer (A) in a thermoplastics constituent preferably 20 – 60 % of the weight, Furthermore, the ethylene and alpha-olefine copolymer of low crystallinity or amorphism nature (B) are to 25 – 40% of the weight, and 1 – 40 % of the weight (C) of ionomers is in 10 – 30% of the weight of a range still more preferably still more preferably 20 to 60% of the weight 25 to 40% of the weight preferably.

[0020] if there is a possibility that heat-sealing reinforcement cannot fully demonstrate [an ethylene vinyl ester copolymer (A)] at less than 20 % of the weight and 60 % of the weight is exceeded on the other hand — voile — there is a possibility that it may be inferior in respect of proper *****. There is a possibility that ethylene and alpha-olefine copolymer (B) cannot demonstrate engine performance sufficient in the field of low-temperature heat-sealing nature and heat-sealing reinforcement at less than 20 % of the weight, and on the other hand, when 60 % of the weight is exceeded, there is a possibility that heat-sealing reinforcement cannot fully demonstrate. There is a possibility that an ionomer (C) cannot demonstrate antistatic effect sufficient at less than 20 % of the weight, and on the other hand, when 60 % of the weight is exceeded, there is a possibility that heat-sealing reinforcement cannot fully demonstrate.

[0021] the thermoplastics constituent of this invention — the three above-mentioned component — adding — a tackifier (D) — content — desirable — 3- if it is made to contain ten to 25% of the weight still more desirable still more preferably and weight % content of will be done 30% of the weight, the stability of the heat-sealing reinforcement as an open-easiness sealant will be improved.

[0022] The thermoplastics constituent of this invention is obtained the ethylene and alpha-olefine copolymer (B) of the above-mentioned ethylene vinyl ester copolymer (A), low crystallinity, or amorphism nature, an ionomer (C), and if needed by the method of mixing a tackifier (D) by the Henschel mixer, V-blender, the ribbon blender, a tumbler mixer, etc. in the above-mentioned range, the method of carrying out melting kneading with a single screw extruder, a multi-screw extruder, a Banbury mixer, etc. further after mixing, etc.

[0023] In case a constituent is separately manufactured, respectively to the ethylene and alpha-olefine copolymer (B) of an ethylene vinyl ester copolymer (A), low crystallinity, or amorphism nature, an ionomer (C), and a tackifier (D), additives, such as an antioxidant usually used, a weathering stabilizer, an antistatic agent, and an antifogger, can be blended with the thermoplastics constituent of this invention in the range which does not spoil the purpose of this invention if needed. The processability and workability in the time of a package etc. can be raised by blending an anti blocking agent and a slipping agent especially at the time of lamination at the time of extrusion.

[0024] As an anti blocking agent, minerals powder, such as a silica, talc, clay, and a zeolite, can be mentioned. as a slipping agent — fatty acids, such as a methylenebis amide of fatty acids, such as fatty-acid amide; stearin acid, such as octadecanamide, oleic amide, a palmitic-acid amide, and an erucic-acid amide, oleic acid, and an erucic acid, and an ethylene bis-amide, — bis— polyalkylene glycol; hydrogenation castor oil; fatty-acid metal salts, such as an amide; polyethylene glycol and a polypropylene glycol, etc. can be raised. As for these anti blocking agent or a slipping agent, it is desirable to blend in the satisfactory range in respect of the engine performance into a thermoplastics constituent.

[0025] The base material (E) by which a laminating is carried out to the sealant of base material (E) this invention usually, it is used as wrapping — various — a material, for example, a polyethylene film, — Polyolefine films, such as a polypropylene film, a polybutene film, and the poly methyl pentene film, Polyester film, such as a polyethylene terephthalate film and a polycarbonate film, A nylon film, a polystyrene film, a polyvinyl chloride film, Films made of thermoplastics, such as a polyvinylidene chloride

film, a polyvinyl alcohol film, an ethylene vinyl alcohol copolymer film, a polymethylmethacrylate film, and ethylene, a vinyl acetate copolymer film, an aluminum foil, paper, etc. are mentioned. The base material which consists of this film made of thermoplastics could be extended, or may be one shaft or a biaxially oriented film. Of course, at least one layer of a base material is good also as more than two-layer.

[0026] The sealant of sealant this invention is the layered product which carried out the laminating of the heat welding layer which becomes at least one side of a base material (E) from the above-mentioned thermoplastics constituent. As a method of manufacturing this sealant, a well-known method, for example, a thermoplastics constituent, is used variously. T-die film making machine, After making it a film beforehand using a tubular blown film making machine etc., The method of sticking without minding through a base material (E) and adhesives, the method of using an extruder for a base material (E), and extruding and laminating a thermoplastics constituent, the method of carrying out co-extrusion shaping of the thermoplastics constituent used as the thermoplastics used as a base material and a heat welding layer, and making it into a sealant, etc. can be illustrated.

[0027] 3-1000 micrometers of thickness of the heat welding layer which consists of a thermoplastics constituent in a sealant are usually in the range of 5-100 micrometers preferably. Moreover, thickness of a heat welding layer can be made thin with about 2 micrometers or more by using the thermoplastics constituent which does not contain the ionomer (C) which is an antistatic agent as a reinforcement layer of a heat welding layer. In that case, the thickness of a reinforcement layer is usually in the range of 18-998 micrometers. A reinforcement layer can also be carried out to one layer or more than two-layer.

[Translation done.]

* NOTICES *

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EXAMPLE

[Example] Next, although an example is given and this invention is explained still more concretely, this invention is not restrained by these examples unless the summary is exceeded.

[0031] The characteristic value in an example and the example of a comparison was measured by the following methods.

(1) Cut down a 10x10cm test piece from the heat welding film obtained the rate of surface specific resistance (aging), and it is in 40-degree C oven, For one day, 30 days, and 90 days, after aging processing, after leaving it from oven for 24 hours in drawing, 20 degrees C, and the ambient atmosphere of 65% of relative humidity, the surface specific resistance value was measured using the digital ultra high resistance by ADVANTEST CORP. / minute amount ammeter (R8340A), and the REJISUCHIBITI chamber (R12704). (Humidity dependency) The 10x10cm test piece was cut down from the obtained heat welding film, and the surface specific resistance value was measured by the above-mentioned method from oven after aging processing for one day in the 60 same% as the relative humidity of drawing and 30-degree C aging processing, 45%, and 35% of ambient atmosphere in 60% of 40-degree-C relative humidity, 45%, and 35% of oven of an ambient atmosphere.

(2) The test piece of A4 size was cut down from the non-charged ***** welding film, and it was left at drawing, 23 degrees C, and 60% of relative humidity from oven after aging processing for one day, 30 days, and 90 days in 40-degree C oven for 24 hours. After having the processed test piece with both hands and rubbing 10 times with the head, it brought close to the ashes of the tobacco which put the test piece on the table, and the distance which ashes were able to draw near to a test piece was measured and evaluated. Even if close brought to 0cm, when the test piece to which the test piece to which ashes do not adhere was brought close to which to 0 and 0-1cm, and ashes adhered only slightly was close brought to 0 and 6cm, the test piece to which ashes adhered was made into x.

(3) Heat-sealing nature : the laminated film was prepared for the field as for which the obtained heat welding film and 12-micrometer biaxial-stretching polyethylene terephthalate film (PET) in thickness carried out the anchor coat with the extrusion lamination process using high pressure process low density polyethylene with a thickness of 20 micrometers. subsequently, superposition and a seal bar 5mm heat sealer were used, and the heat welding film plane of a laminated film and the polypropylene sheet (TOH CELLO PP sheet T-T) side with a thickness of 300 micrometers which were acquired were heat sealed for 1 second by the pressure of 0.2MPa with a predetermined temperature — it cooled radiationally. From now on, the test piece of 15mm width of face was cut off, the heat-sealing section was exfoliated in a part for 500mm/in crosshead speed, and the reinforcement was made into heat-sealing reinforcement.

[0032] Moreover, the raw material used by this example is as follows.

(1) ethylene-vinylacetate copolymer (EVA): — MFR4.0g / [10 % of the weight of (2) ethylene and 1-butene copolymer (EBR):1-butene contents with 10 % of the weight of vinyl acetate contents, 10 minutes / MFR9g / minutes /, and a melting point of 94 degrees C, density 0.88 g/cm³, the melting point of 68 degrees C, and] 115 degrees-C (4) glycerol of 10-minute aliphatic-series [(3)] system hydrocarbon resin:Arakawa Chemistry product (trade name Al Cong) and ring and ball method softening temperatures 10wt(s)% — the ethylene acrylic-acid copolymer potassium ionomer resin constituent (ionomer constituent) to contain

Base polymer: Ethylene acrylic-acid copolymer (21 % of the weight of acrylic-acid contents)

source of metal cation: — potassium neutralization whenever: — 80 mol % metal cation content: — 2.3-mol [/kg] ionomer MFR:0.6g /, and 10-minute glycerol (molecular weight 92)

[0033] Example 1EVA: Ionomer [36 % of the weight, EBR:36 % of the weight, aliphatic series system hydrocarbon resin:18 % of the weight, and] constituent: The thermoplastic constituent (heat welding layer) which carried out the dryblend of the 10 % of the weight was supplied to the extruder, and the heat welding film was obtained by the T-die method. The thickness of a film was 30 micrometers. The evaluation result of the obtained heat welding film is shown in a table 1.

[0034] Example 2EVA: Ionomer [34 % of the weight, EBR:34 % of the weight, aliphatic series system hydrocarbon resin:17. % of the weight, and] constituent: The thermoplastic constituent (heat welding layer) which carried out the dryblend of the 15 % of the weight was supplied to the extruder, and the heat welding film was obtained by the T-die method. The thickness of a film was 30 micrometers. The evaluation result of the obtained heat welding film is shown in a table 1.

[0035] Example 3EVA: Ionomer [32 % of the weight, EBR:32 % of the weight, aliphatic series system hydrocarbon resin:16 % of the weight, and] constituent: The thermoplastic constituent (heat welding layer) which carried out the dryblend of the 20 % of the weight was supplied to the extruder, and the heat welding film was obtained by the T-die method. The thickness of a film was 30 micrometers. The evaluation result of the obtained heat welding film is shown in a table 1.

[0036] Example 4EVA: Ionomer [40 % of the weight, EBR:40 % of the weight, and] constituent: The thermoplastic constituent (heat welding layer) which carried out the dryblend of the 20 % of the weight was supplied to the extruder, and the heat welding film was obtained by the T-die method. The thickness of a film was 30 micrometers. The evaluation result of the obtained heat welding film is shown in a table 1.

[0037] Aliphatic-series [example of comparison 1EVA:40 % of the weight, EBR:40 % of the weight, and] system hydrocarbon resin: The thermoplastic constituent (heat welding layer) which carried out the dryblend of the 20 % of the weight was supplied to the extruder, and the heat welding film was obtained by the T-die method. The thickness of a film was 30 micrometers. The evaluation result of the obtained heat welding film is shown in a table 1.

[0038]

[A table 1]

評価項目	例1	例2	例3	例4	比較例1
引張強度					
140℃	8.0	1.5	8.0	8.0	7.5
160℃	8.8	8.8	8.8	8.0	9.0
180℃	12.6	12.6	12.0	10.0	12.5
伸び率					
40℃×10	1.8×10 ⁻³	8.0×10 ⁻³	8.2×10 ⁻³	1.8×10 ⁻³	2.8×10 ⁻³
100℃×300	2.0×10 ⁻³	1.0×10 ⁻³	0.1×10 ⁻³	0.8×10 ⁻³	2.3×10 ⁻³
40℃×300	2.8×10 ⁻³	2.0×10 ⁻³	0.8×10 ⁻³	1.5×10 ⁻³	6.8×10 ⁻³
重量減少率					
0.07%	—	—	2.1×10 ⁻³	3.0×10 ⁻³	0.5×10 ⁻³
4.52%	—	—	1.8×10 ⁻³	0.7×10 ⁻³	0.5×10 ⁻³
3.52%	—	—	0.5×10 ⁻³	1.0×10 ⁻³	0.0×10 ⁻³
全長変化					
40℃×10	0~0	0	0	0	0
100℃×300	0~0	0	0	0	0
40℃×300	0~0	0	0	0	0

[Translation done.]